The Chemistry on Diterpenoids in 1981 (Commemoration Issue Dedicated to Professor Masaya Okano on the Occasion of his Retirement)

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REVIEW

The Chemistry on Diterpenoids in 1981

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Manabu NODE*, and Masahito OCHIAI*

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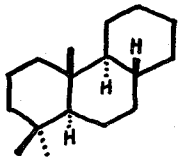
I. INTRODUCTION

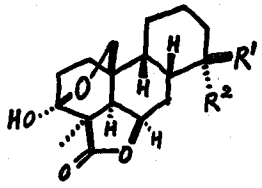
This is one of a series of our annual reviews on diterpenoid chemistry. The following abbreviations are used. [CN]: common name; [NS]: natural source; [REF]; reference number; [NC]: notes and comments.

II. PODOCARPANE DERIVATIVES

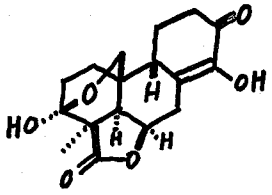


Podocarpane

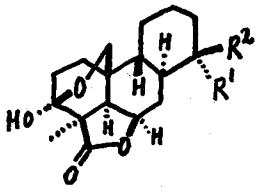
1) Isolation and Structure Determination



1 $R^1+R^2=0$
[CN] humirianthenolide A
2 $R^1=H$, $R^2=OH$
[CN] humirianthenolide B
[NS] *Humirianthera rupestris*
[REF] 1



3
[CN] humirianthenolide D
[NS] *Humirianthera rupestris*
[REF] 1



4 $R^1+R^2=0$
[CN] humirianthenolide E
5 $R^1=H$, $R^2=OH$
[CN] humirianthenolide F
[NS] *Humirianthera rupestris*
[REF] 1

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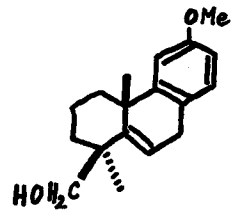
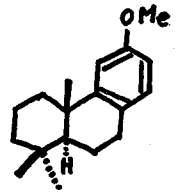
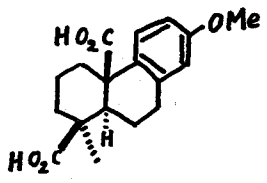
2) Synthesis and Reaction



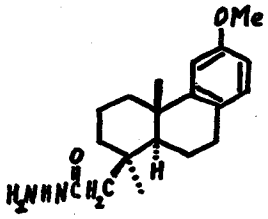
6
[REF] 2
[NC] chemical conversion



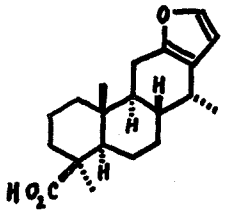
7
[REF] 3
[NC] synthesis



8
[REF] 4
[NC] synthesis

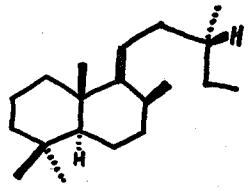


9
[REF] 5
[NC] oxidation



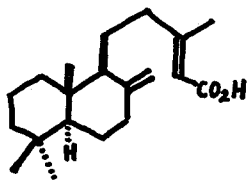
10
[CN] (+)-methyl vouacapenate
[REF] 6
[NC] synthesis from podocarpic acid

III. LABDANE DERIVATIVES

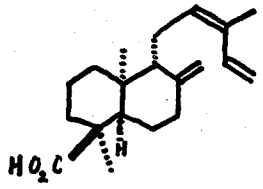


Labdane

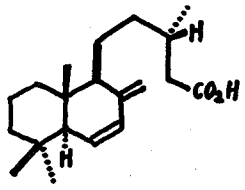
1) Isolation and Structure Determination



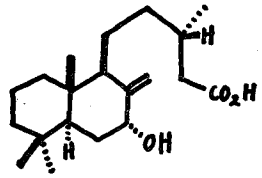
11
[NS] *Eperua purpurea*
[REF] 7



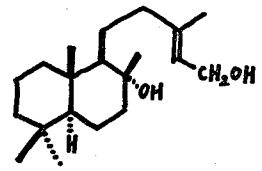
12
[CN] *cis*-ozic acid
[NS] *Helianthus angustifolius*
[REF] 8



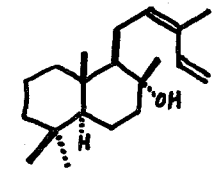
13
[CN] cistadienic acid
[NS] *Cistus symphytifolius*
[REF] 9



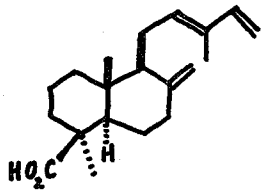
14
[CN] cistenolic acid
[NS] *Cistus symphytifolius*
[REF] 9



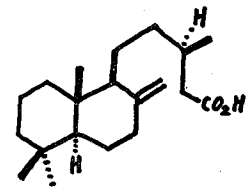
15
[NS] *Cistus symphytifolius*
[REF] 9



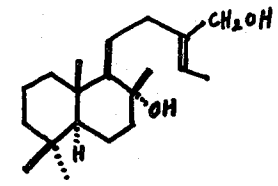
16
[CN] (+)abienol
[NS] *Fleischmannia pycnocephaloides*
[REF] 10



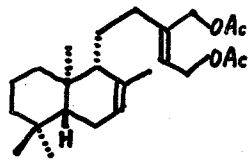
17
[NS] *Fleischmannia deborabellae*
[REF] 10



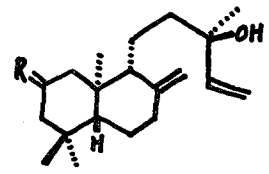
18
[NS] *Eperua purpurea*
[REF] 11



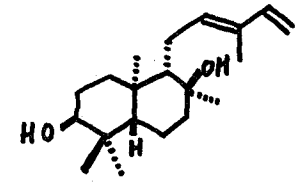
19
[NS] *Picea ajanensis*
[REF] 12



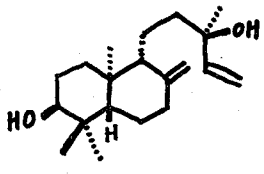
20
[NS] *Baccharis* species
[REF] 13



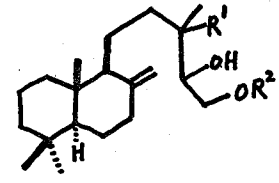
21 R=α-OH, H
22 R=β-OH, H
23 R=0
24 R=α-OCO$(CH_2)_2CO_2H$, H
[NS] *Baccharis oxydonta*
[REF] 14



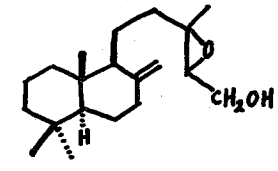
25
[NS] *Grazielia* species
[REF] 15



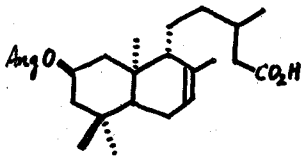
26
[NS] *Croton sublyratus*
[REF] 16



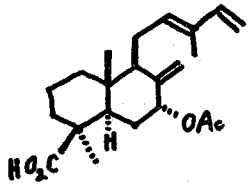
27 R^1=R^2=H
28 R^1=OH, R^2=H
29 R^1=OH, R^2=Ac
[NS] *Hemizonia lutescens*
[REF] 17



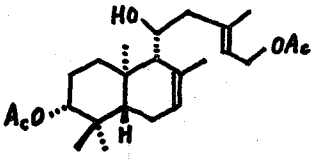
30
[NS] *Hemizonia lutescens*
[REF] 17



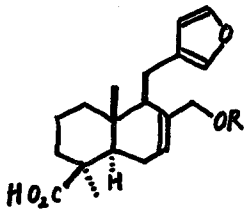
31
[CN] dihydrodendroidinic acid
[NS] *Pleurocoronis pluriseta*
[REF] 18



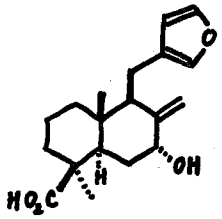
32
[NS] *Chromolaena collina*
[REF] 19



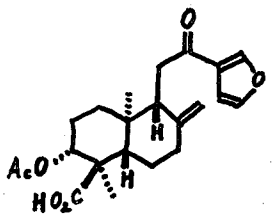
33
[NS] *Lasiolaena santosii*
[REF] 20



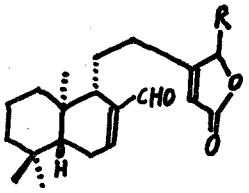
34 R=H
35 R=Ac
[NS] *Gutierrezia dracunculoides*
[REF] 21



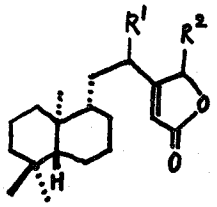
36
[NS] *Gutierrezia dracunculoides*
[REF] 21



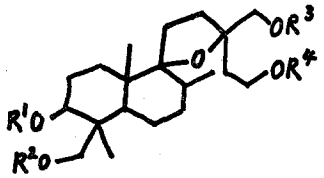
37
[NS] *Dodonaea petiolaris*
[REF] 22



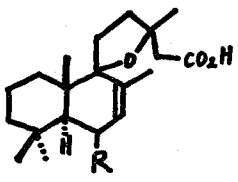
38 R=β-OH
[CN] acritolongifolide A
39 R=α-OH
[CN] acritolongifolide B
[NS] *Acritopappus longifolius*
[REF] 23



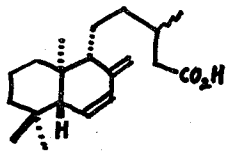
40a R^1=H, R^2=α-OH
40b R^1=H, R^2=β-OH
41a R^1=OH, R^2=α-OH
41b R^1=OH, R^2=β-OH
[NS] *Ageratum fastigiatum*
[REF] 24



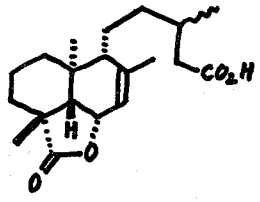
42 R^1=R^3=Ac, R^2=R^4=H
43 R^1=R^4=Ac, R^2=R^3=H
44 R^1=R^3=H, R^2=R^4=Ac
[NS] *Lagochilus inebrians*
[REF] 25



45 R=α-OH
46 R=β-OH
[NS] *Grindelia humilis*
[REF] 26

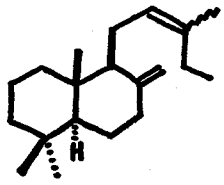


47
[NS] *Hartwrightia floridana*
[REF] 27

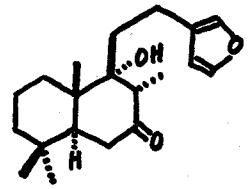


48
[NS] *Hartwrightia floridana*
[REF] 27

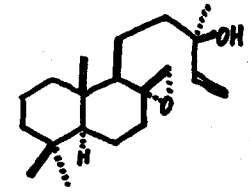
2) Synthesis and Reactions



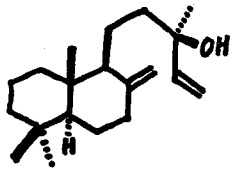
49
[REF] 28
[NC] photosensitized oxygenation of **49** and related compds.



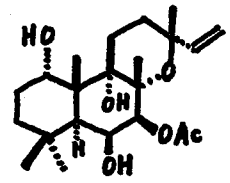
50
[CN] hispanolone
[REF] 29, 30, 31
[NC] chemical transformation of **50**



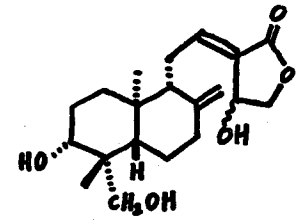
51
[REF] 32
[NC] acid-catalysed rearrangement



52
[CN] manool
[REF] 33
[NC] conversion to isoagatholactone

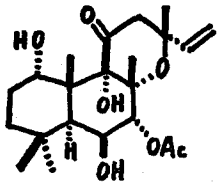


53
[CN] forskolin
[NS] *Coleus forskohlii*
[REF] 34
[NC] reactions

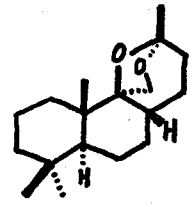


54
[CN] andrographolide
[REF] 35
[NC] reaction with $NaHSO_3$

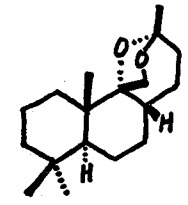
3) Miscellaneous Section



55
[CN] coleonol
[NS] *Coleus forskohlii*
[REF] 36
[NC] pharmacologial studies



56
[REF] 37
[NC] •synthesis from manool
•odour studies



57
[REF] 37
[NC] •synthesis from manool
•odour studies

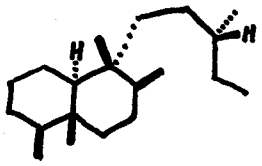
Additional references

[REF] 38 - 41
[NC] ^{13}C NMR studies

[REF] 42
[NC] studies on optical activity

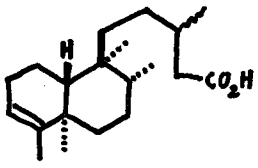
[REF] 43
[NC] separation by HPLC

IV. CLERODANE DERIVATIVES

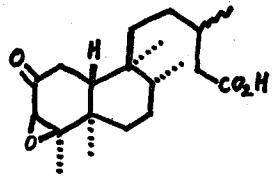


Clerodane

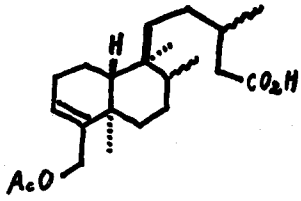
1) Isolation and Structure Determination



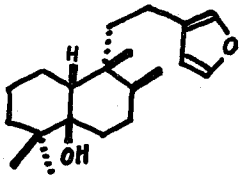
58
[NS] *Hartwrightia floridana*
[REF] 27



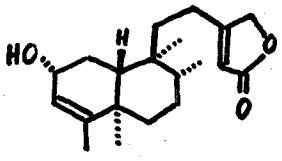
59
[NS] *Hartwrightia floridana*
[REF] 27



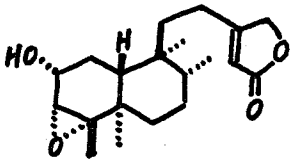
60
[NS] *Liatris scariosa*
[REF] 44



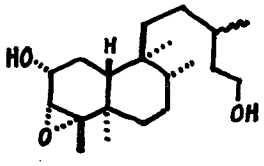
61
[NS] *Dysidea ambia*
[REF] 45



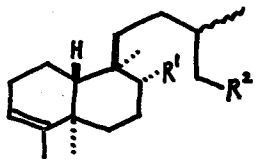
62
[NS] *Symphyopappus compressus*
[REF] 46



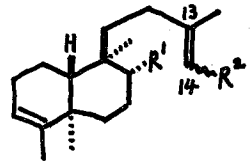
63
[NS] *Symphyopappus compressus*
[REF] 46



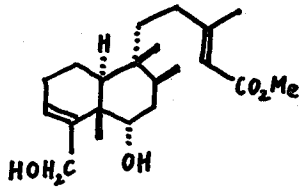
64
[NS] *Symphyopappus compressus*
[REF] 46



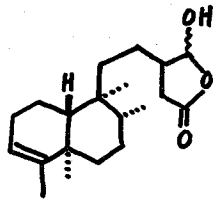
65 $R^1=CH_2OH$, $R^2=CHO$
66 $R^1=CH_2OH$, $R^2=CH_2OH$
67 $R^1=CH_2OH$,
$R^2=CH_2OCO(CH_2)_{18}Me$
68 $R^1=CH_2OH$,
$R^2=CH_2OCO(CH_2)_{20}Me$
69 $R^1=CHO$, $R^2=CH_2OH$
[NS] *Symphyopappus reticulatus*
[REF] 46



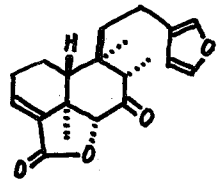
70 $R^1=CH_2OH$, $R^2=CHO$
(13, 14 E and Z)
71 $R^1=CO_2H$, $R^2=CHO$
(13, 14 E and Z)
72 $R^1=R^2=CH_2OH$ (13, 14 E)
73 $R^1=CH_2OH$,
$R^2=CH_2OCO(CH_2)_{18}Me$ (13, 14 E)
74 $R^1=CH_2OH$,
$R^2=CH_2OCO(CH_2)_{20}Me$ (13, 14 E)
[NS] *Symphyopappus reticulatus*
[REF] 46



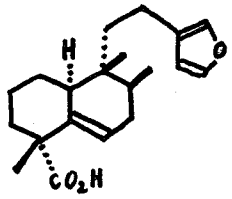
75
[NS] *Pityrodia lepidota*
[REF] 47



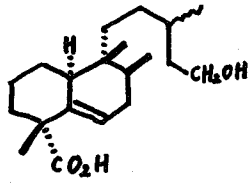
76
[NS] *Balianthus viscidus*
[REF] 48



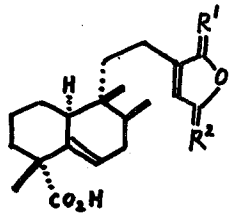
77
[NS] *Pulicaria gnaphalodes*
[REF] 49



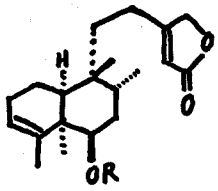
78
[CN] koanophyllic acid A
[NS] *Koanophyllon conglobatum*
[REF] 50



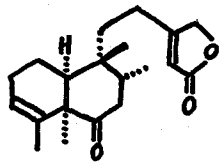
79
[CN] koanophyllic acid B
[NS] *Koanophyllon conglobatum*
[REF] 50



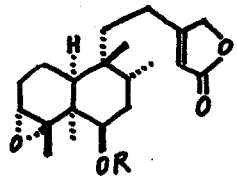
80 $R^1=O$, $R^2=H_2$
[CN] koanophyllic acid C
81 $R^1=H_2$, $R^2=O$
[CN] koanophyllic acid D
[NS] *Koanophyllon conglobatum*
[REF] 50



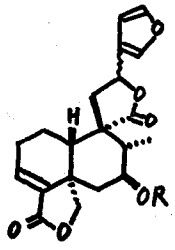
82 R=angeloyl
[CN] solidagolactone II
83 R=tigloyl
[CN] solidagolactone III
84 R=H
[CN] solidagolactone IV
85 R=Ac
[CN] solidagolactone VIII
[REF] 51
[NC] structure revision



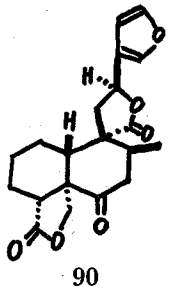
86
[CN] solidagolactone V
[REF] 51
[NC] structure revision



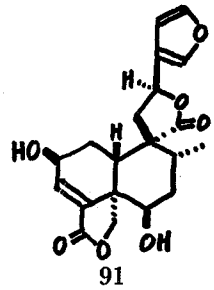
87 R=Ac
[NC] solidagolactone VI
88 R=angeloyl
[CN] solidagolactone VII
[REF] 51
[NC] structure revision



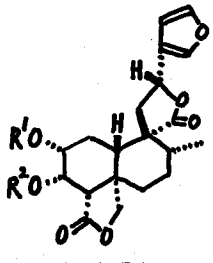
89 R=angeloyl
[NS] *Baccharis subdentata*
[REF] 14



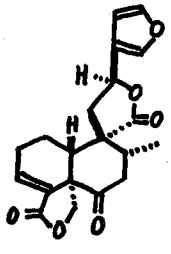
90
[NS] *Teucrium scordium*
[REF] 52



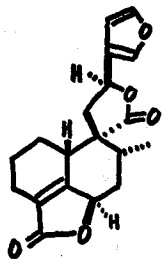
91
[CN] teugin
[NS] *Teucrium fragile*
[REF] 53



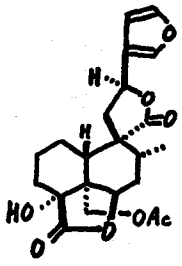
92 R^1=angeloyl, R^2=senecioyl
93 R^1=senecioyl, R^2=angeloyl
94 R^1=angeloyl, R^2=2-methylbutyl
[NS] *Baccharis* species
[REF] 13



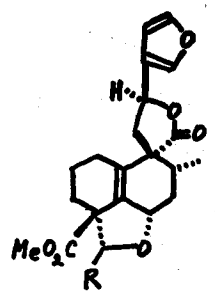
95
[CN] teuscordinon
[NS] *Teucrium scordium*
[REF] 54



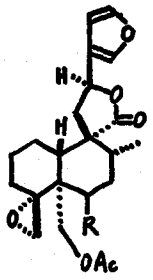
96
[CN] teuflin
[NS] *Teucrium viscidum var. Miguelianum*
[REF] 55



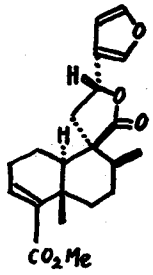
97
[NS] *Teucrium* species
[REF] 56



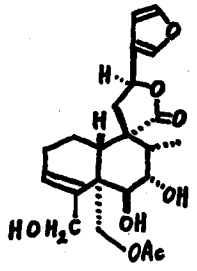
98 R=β-OH
[CN] mallotucin C
99 R=α-OH
[CN] mallotucin D
[NS] *Mallotus repandus*
[REF] 57



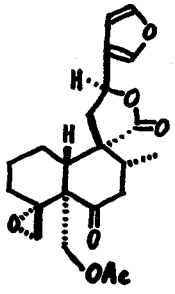
100 R=β-OH
[CN] teucjaponin A
101 R=α-OH
[CN] teucjaponin B
[NS] *Truvtium japonicum*
[REF] 58



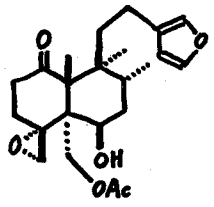
102
[CN] sonderianin
[NS] *Croton sonderianus*
[REF] 59



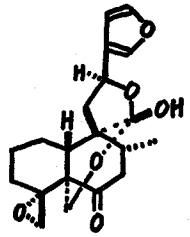
103
[CN] lolin
[NS] *Teucrium capitatum*
[REF] 60



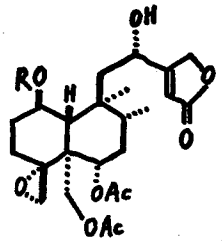
104
[NS] *Teucrium gnaphalodes*
[REF] 61



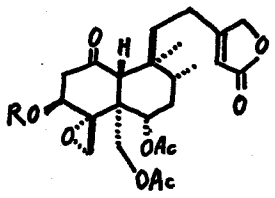
105
[CN] isofruticolone
[NS] *Teacrium gnaphalodes*
[REF] 61



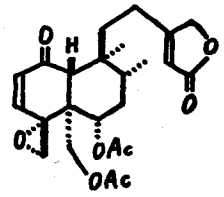
106
[NS] *Teucrium gnaphalodes*
[REF] 61



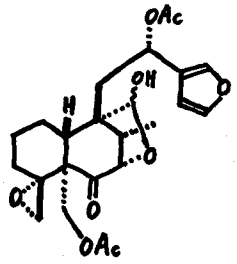
107 R=COC(Me)=^{E}CHMe
[CN] ajugamarin
[NS] *Ajuga nipponensis*
[REF] 62



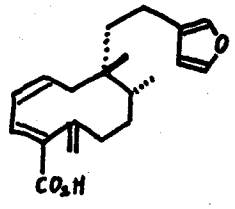
108 R=COCH(Me)Et
[CN] ajugareptansone A
[NS] *Ajuga reptans*
[REF] 63



109
[CN] ajugareptansone B
[NS] *Ajuga reptans*
[REF] 63

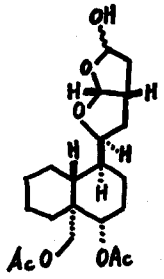


110
[CN] auropolin
[NS] *Teacrium polium* snbsp. *aureum*
[REF] 64

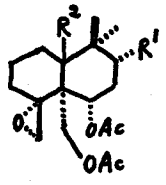


111
[CN] conyzic acid
[NS] *Conyza strict*
[REF] 65

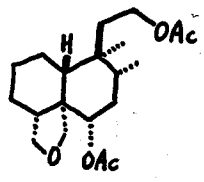
2) synthesis and Reactions



112
[REF] 66
[NC] ·synthesis
·insect antifeedant



113 R^1=H, R^2=β-H
114 R^1=H, R^2=α-H
115 R^1=Me, R^2=β-H
[REF] 67 - 69
[NC] ·synthesis
·insect antifeedant



116
[REF] 70
[NC] synthesis

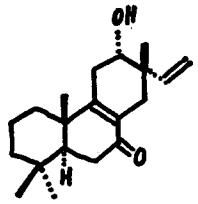
3) Miscellaneous Section

[REF] 71
[NC] studies on insect antifeeding activity

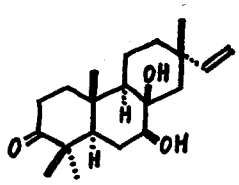
V. PIMARANE AND ISOPIMARANE DERIVATIVES

Pimarane and Isopimarane

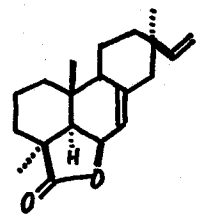
1) Isolation and Structure Determination



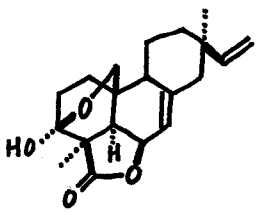
117
[NS] *Vellozia* species
[REF] 72



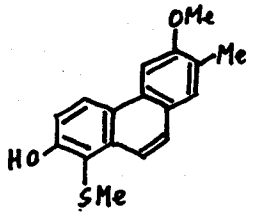
118
[NS] *Bromelia pinguin*
[REF] 73



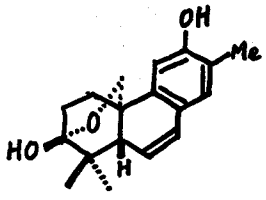
119
[CN] momilactone A
[NS] rice
[REF] 74
[NC] phytoalexin



120
[CN] momilactone B
[NS] rice
[REF] 74
[NC] phytoalexin

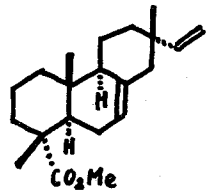


121
[CN] micrandrol C
[NS] *Micrandropsis scleroxylon*
[REF] 75



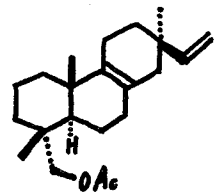
122
[CN] micrandrol D
[NS] *Micrandropsis scleroxylon*
[REF] 75

2) Synthesis and Reaction



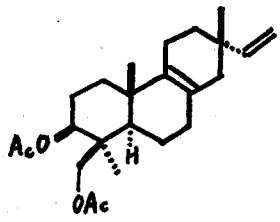
123

[REF] 76
[NC] superacid catalyzed cyclization



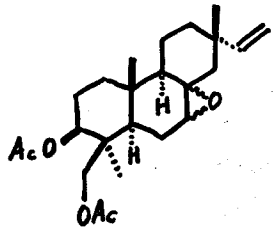
124

[REF] 77
[NC] photo-oxygenation



125

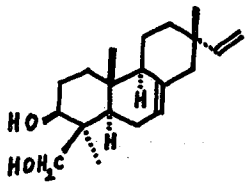
[REF] 77
[NC] photo-oxygenation



126

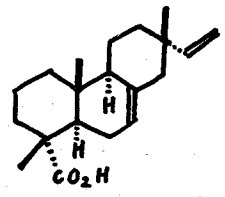
[REF] 78
[NC] chemical transformation

3) Micsellaneous Section



127

[CN] virescenol B
[REF] 79
[NC] bioxynthesis



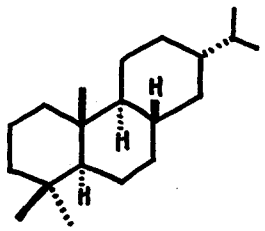
128

[CN] isopimaric acid
[REF] 80
[NC] biotransformation

Additional references

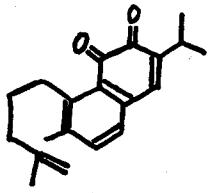
[REF] 81 - 83
[NC] ^{13}C NMR sutdies
[REF] 84
[NC] studies on distribution of podolactone-type plant growth inhibitors
[REF] 85
[NC] studies on conformations for pimarane derivatives

VI. ABIETANE DERIVATIVES

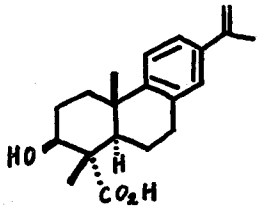


Abietane

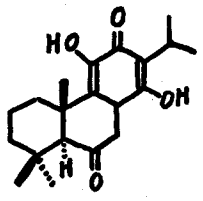
1) Isolation and Structure Determination



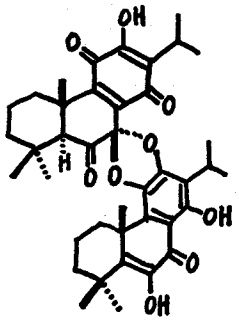
129
[CN] aethiopinone
[NS] *Salvia aethiopis*
[REF] 86



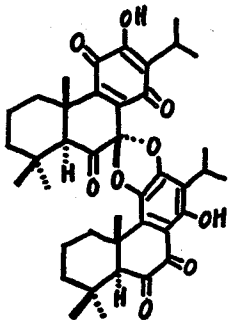
130
[NS] *Salvia tomentosa*
[REF] 87



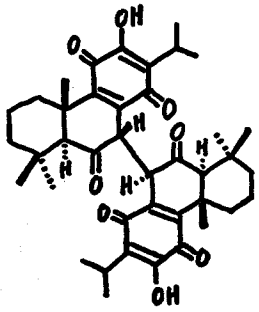
131
[NS] *Plectranthus grandidentatus*
[REF] 88



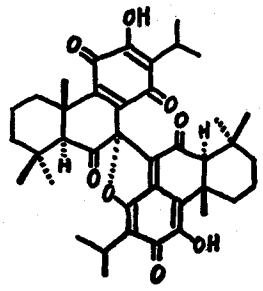
132 [CN] grandidone A
133 [CN] 7-epigrandidone A
[NS] *Plectranthus grandidentatus*
[REF] 88



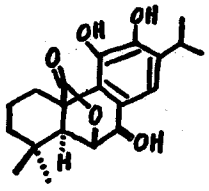
134 [CN] grandidone B
135 [CN] 7-epigrandidone B
[NS] *Plectrantus grandidentatus*
[REF] 88



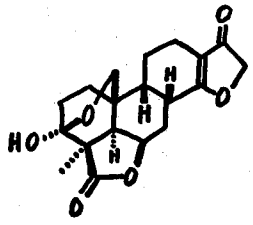
136
[CN] grandidone C
[NS] *Plectrantuhs grandidentatus*
[REF] 88



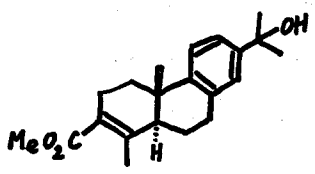
137 [CN] grandidone D
138 [CN] 7-epigrandidone D
[NS] *Plectranthus grandidentatus*
[REF] 88



139
[CN] rosmanol
[NS] *Rosmarinus officinalis*
[REF] 89
[NC] antioxidant

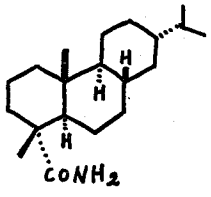


140
[CN] humiliathenolide C
[NS] *Humirianthera rupestris*
[REF] 1

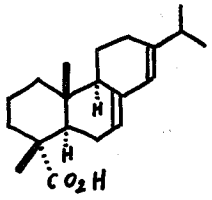


141
[NS] *Tripterygium wilfordii*
[REF] 90
[NC] isolation of cytotoxic diterpents. (tripdioloide, triptolide, etc.)

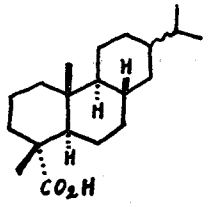
2) Synthesis and Reaction



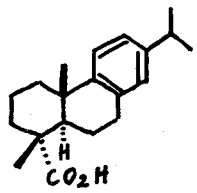
142
[REF] 91
[NC] lactonization by photolysis ($Pb(OAc)_4/I_2$)



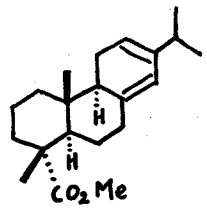
143
[CN] abietic acid
[REF] 92
[NC] air oxidation



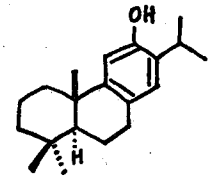
144
[REF] 93
[NC] carbonylation by conc. H_2SO_4



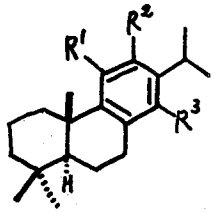
145
[CN] dehydroabietic acid
[REF] 94
[NC] oxidation by $Co(acac)_2$
[REF] 95
[NC] Synthesis of (+)-fragrolide and (+)-bemadienolide



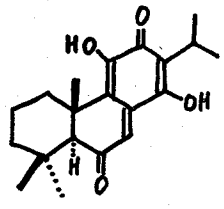
146
[CN] methy levopimarate
[REF] 96
[NC] addition of $ClSO_2NCO$



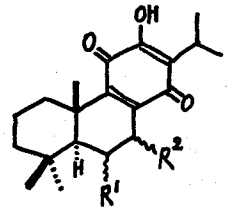
147
[CN] ferruginol
[REF] 97
[NC] synthesis from **145**
[REF] 98
[NC] ozonolysis



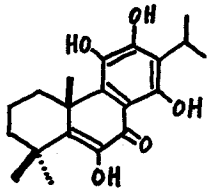
148 $R^1=R^2=OH$, $R^3=H$
149 $R^1=R^2=H$, $R^3=OH$
[REF] 98
[NC] Chemical conversion to isodrimenin, validivioloide, winterin, and confertifolin



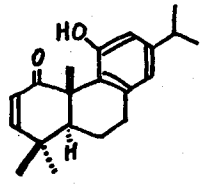
150
[CN] 14-hydroxytaxodione
[REF] 99
[NC] partial synthesis and reactions of **150**



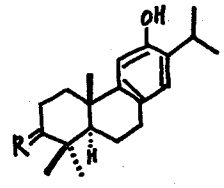
151 $R^1=R^2=\alpha$-OH
152 $R^1=\alpha$-OH, $R^2=\beta$-OH
153 $R^1=R^2=\beta$-OH
[REF] 100
[NC] partial synthesis



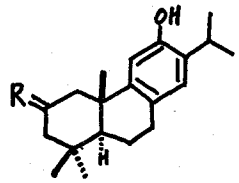
154
[CN] coleoneU
[REF] 101 and 102
[NC] synthesis



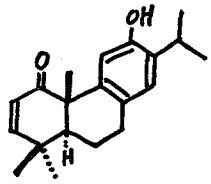
155
[REF] 103
[NC] synthesis



156 R=α-H, β-OH
[CN] hinokiol
157 R=O
[CN] hinokione
[REF] 104
[NC] synthesis from abietatrienol
[REF] 105
[NC] total synthesis



158 R=α-OH, β-H
[CN] salviol
159 R=O
[CN] 2-oxoferruginol
[REF] 104
[NC] synthesis from abietatrienol
[REF] 105
[NC] total synthesis

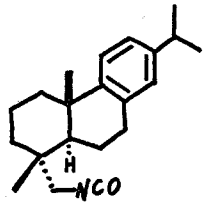


160
[CN] shonanol
[REF] 106
[NC] synthesis and revised structure

131
[REF] 107
[NC] partial synthesis (**131-138**) from **131**

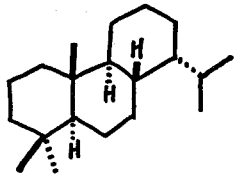
3) Miscellaneous Section

145
[REF] 108
[NC] biodegradation of dehydroabietic acid (**145**) with *Mortierella isabellina*



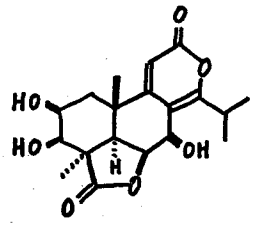
161
[REF] 109
[NC] use in the synthesis of (*S*)-5-hydroxy-6-*trans*-8, 11, 14-*cis*-eicosatetraenoic acid

VII. TOTARANE DERIVATIVES



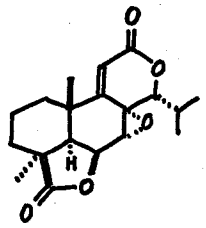
Totarane

1) Isolation and Structure Determination



162

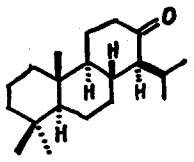
[CN] urbalactone
[NS] *Podocarpus urbanii*
[REF] 110



163

[CN] 2,3-dihydropodolide
[NS] *Podocarpus urbanii*
[REF] 110

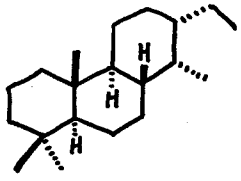
2) Synthesis and Reaction



164

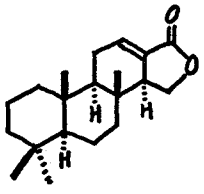
[REF] 111
[NC] Synthesis *via* cyclobutane ring-opening of allene-enone photoadduct

VIII. CASSANE DERIVATIVES



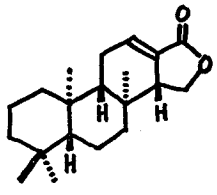
Cassane

1) Synthesis and Reaction



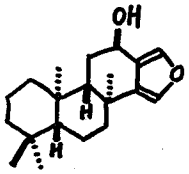
165

[CN] (+)-isoagatholactone
[REF] 33
[NC] synthesis from (+)-manool

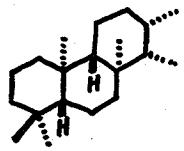


166

[CN] *ent*-isoagatholactone
[REF] 112
[NC] synthesis from methyl isocopalate

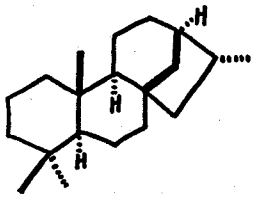


167
[CN] *ent*-13 (16),14-spongiadien-12α-ol
[REF] 112
[NC] synthesis from methyl isocopalate



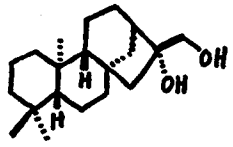
168
[CN] isocopalane
[REF] 112
[NC] synthesis

IX. KAURANE DERIVATIVES

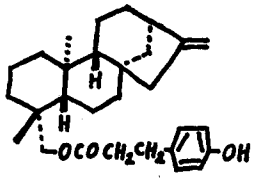


Kaurane

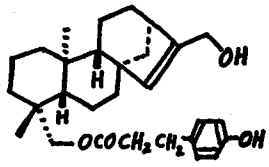
1) Isolation and Structure Determination



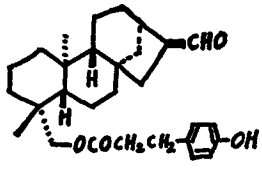
169
[NS] *Croton sublyratus*
[REF] 16
[NS] *Aristolachia elegans*
[REF] 113



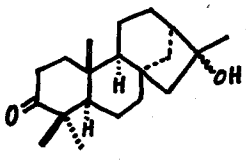
170
[NS] *Baccharis quitensis*
[REF] 14



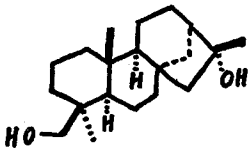
171
[NS] *Baccharis quitensis*
[REF] 14



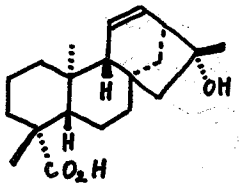
172
[NS] *Baccharis quitensis*
[REF] 14



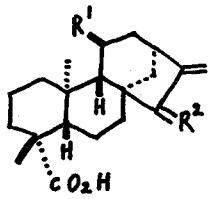
173
[NS] *Bromelia pinguin*
[REF] 73



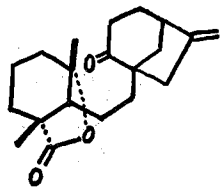
174
[NS] *Bromelia pinguin*
[REF] 73



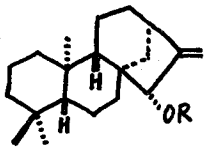
175
[NS] *Helianthus angustifolius*
[REF] 8



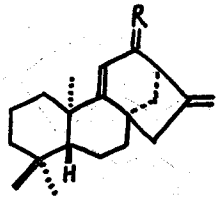
176 R^1=H, R^2=α-OTigl
177 R^1=OH, R^2=O
178 R^1=OH, R^2=β-OH, H
[NS] *Grazielia* species
[REF] 15



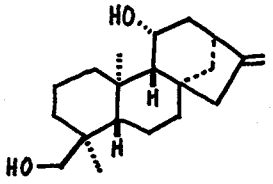
179
[NS] *Ageratum fastigiatum*
[REF] 24



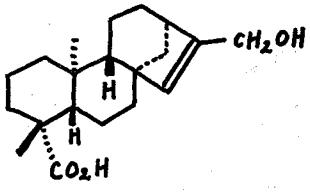
R=methylacryloyl
180
[NS] *Ichthyothere* species
[REF] 114



181 R=α-H, β-OH
182 R=O
[NS] *Vellozia caput-ardeae*
[REF] 115

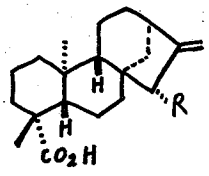


183
[NS] *Sideritis arborescens*
[REF] 116



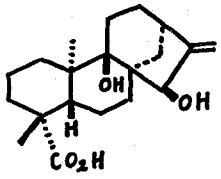
184

[NS] *Helianthus grosseserratus*
[REF] 117

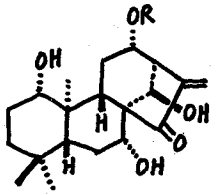


185 R=OH
[CN] grandifloric acid
[NS] *Helianthus grosseserratus*
[REF] 117
186 R=H
[NS] *Aristolochia triangularis*
[REF] 118
[NS] *Wedelia buphthalmiflora*
[REF] 119

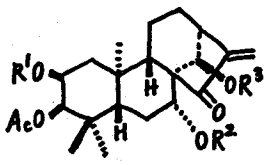
187 R=H; $\Delta^{9(11)}$
[CN] grandiflorenic acid
188 R=MeCH=CMeCOO-(Z)
189 R=MeCH=CMeCOO-(E)
190 R=Me_2CHCH_2COO-
191 R=Me_2CHCOO-
[NS] *Wedelia buphthalmiflora*
[REF] 119



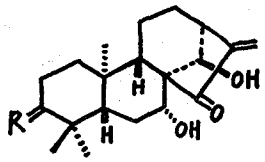
192
[CN] pterokaurene L_2
[NS] *Pteris longipes*
[REF] 120



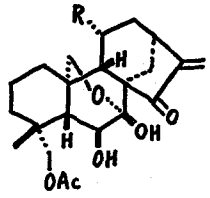
193 R=H
[CN] excisanin A
194 R=Ac
[CN] excisanin B
[NS] *Rabdosia excisa*
[REF] 121



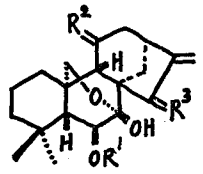
195 $R^1=R^2=R^3=H$
[CN] leukamenin A
196 R^1=Ac, $R^2=R^3=H$
[CN] leukamenin B
197 $R^1=R^3$=Ac, $R^2=H$
[CN] leukamenin C
198 $R^1=R^2$=Ac, $R^3=H$
[CN] leukamenin D
[NS] *Rabdosia umbrosa var. leucantha f. kameba*
[REF] 122



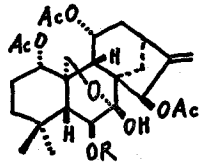
199 R=α-H, β-OAc
[CN] leukamenin E
200 R=O
[CN] leukamenin F
[NS] *Rabdosia umbrosa var. leucantha f. kameba*
[REF] 122



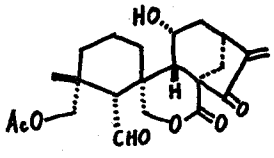
201 R=H
[CN] longikaurin C
202 R=OH
[CN] longikaurin D
203 R=OAc
[CN] longikaurin F
[NS] *Rabdosia longituba*
[REF] 123
[NC] antibacterial diterpenoids



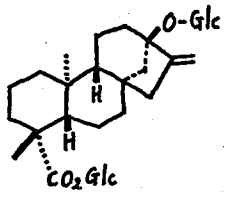
204 R^1=H, R^2=α-H, β-OAc, R^3=O
[CN] longikaurin E
[NS] *Rabdosia longituba*
[REF] 123
205 R^1=OAc, R^2=O, R^3=α-H, β-OH
[CN] rabdosianin C
[NS] *Rabdosia shikokiana*
[REF] 124



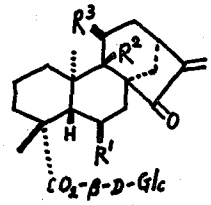
206 R=H
[CN] rabdosianin A
207 R=Ac
[CN] rabdosianin B
[NS] *Rabdosia shikokiana*
[REF] 124



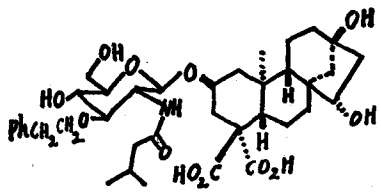
208
[CN] trichorabdal B
[NS] *Rabdosia trichocarpa*
[REF] 125
[NC] X-ray crystal structure and transformation into novel skeleton



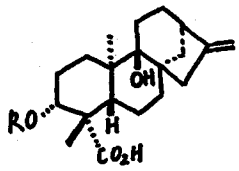
209
[CN] rubusoside
[NS] *Rubus chingii*
[REF] 126
[NC] sweet principle



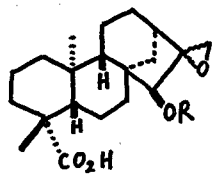
210 $R^1=R^3=H$, R^2=OH
211 $R^1=R^3$=OH, $R^2=H$
212 $R^1=R^2$=OH, $R^3=H$
[NS] *Pteris livida*
[REF] 127



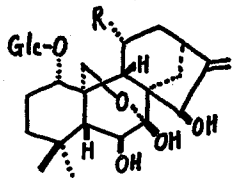
213
[CN] wedeloside
[NS] *Wedelia asperrima*
[REF] 128 and 129
[NC] toxicity and potential antitumor activity



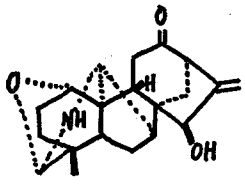
214 R=Ang
215 R=Cinn
[NS] *Wedelia trilobata*
[REF] 130



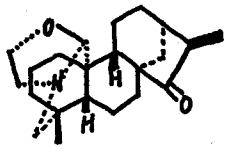
216 R=Ang
217 R=Tigl
[NS] *Aspilia parvifolia*
[REF] 130



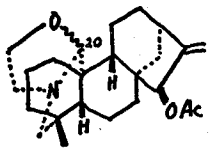
218 R=OH
[CN] shikokiaside A
219 R=H
[CN] shikokiaside B
[NS] *Rabdosia shikokiana var. shikokiana*
[REF] 131



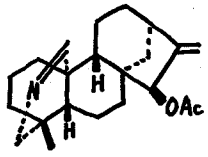
220
[CN] norsongoramine
[NS] *Delphinium tamarae*
[REF] 132



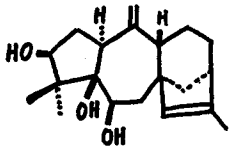
221
[CN] cuauchichicine
[REF] 133
[NC] revised structure and ^{13}C-NMR and X-ray crystallography



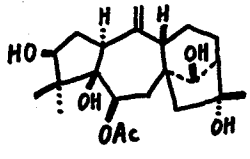
222
[CN] ovatine
[NS] *Garrya ovata var. lindheimeri*
[REF] 133
[NC] mixture of C(20) epimer



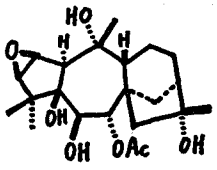
223
[CN] lindheimerine
[NS] *Garrya ovata var. lindheimeri*
[REF] 133



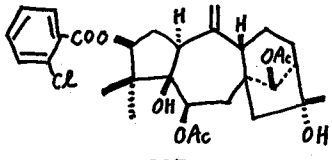
224
[CN] grayanotoxin XIX
[NS] *Leucothoe grayana*
[REF] 134



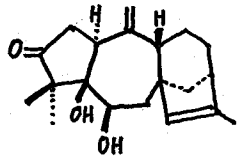
225
[CN] grayanotoxin XVI
]NS] *Leucothoe grayana*
[REF] 135



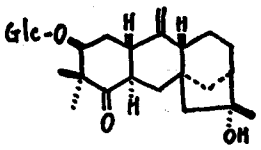
226
[CN] lyoniatoxin
[NS] *Lyonia ovalifolia var. elliptica*
[REF] 136
[NC] revised structure



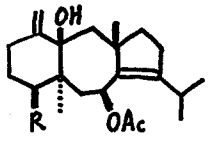
227
[REF] 137
[NC] X-ray analysis and boat conformation of C-ring



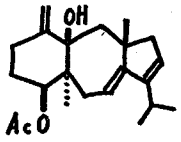
228
[CN] grayanotoxin XVIII
[NS] *Leucothoe grayana*
[REF] 138
[NC] X-ray analysis



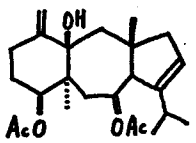
229
[CN] pieroside B
[NS] *Pieris japonica*
[REF] 139
[NC] first example of a leucothane glycoside



230 R=H
231 R=OH
[NS] *Dictyota divaricata*
[REF] 140
[NC] X-ray analysis

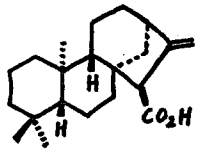


232
[NS] *Dictyota divaricata*
[REF] 140
[NC] X-ray analysis

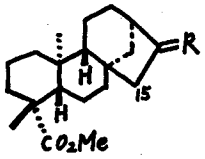


233
[NS] *Dictyota divaricata*
[REF] 140
[NC] X-ray analysis

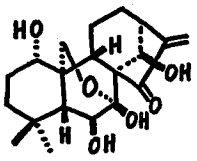
2) Synthesis and Reaction



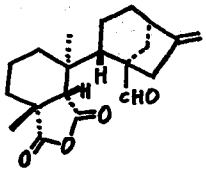
234
[REF] 141
[NC] carboxylation of kaurene



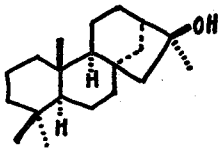
235 R=F_2
236 R=CF_3 (15-en)
[REF] 142
[NC] derived from xylopic acid



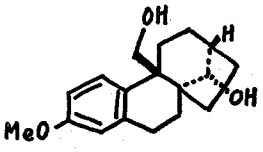
237
[CN] oridonin
[REF] 143
[NC] selective acetylation



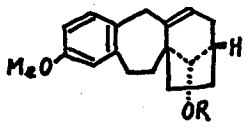
238
[CN] fujenal
[REF] 144 and 145
[NC] preparation of some 7-norgibberellanes from **238**



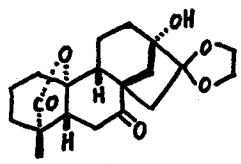
239
[CN] phyllocladan-16β-ol
[REF] 146
[NC] synthesis from phyllocladene



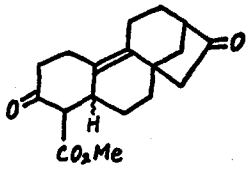
240
[REF] 147
[NC] synthetic approach to grayanotoxins



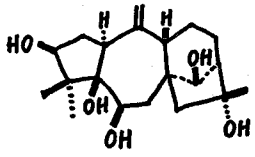
241
[REF] 148
[NC] synthetic approach to grayanotoxins



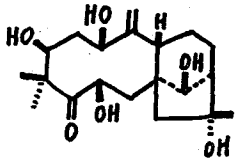
242
[REF] 149
[NC] synthetic approach to gibberellins



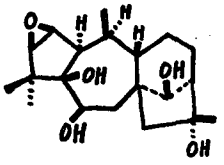
243
[REF] 150
[NC] synthetic approach to gibberellins



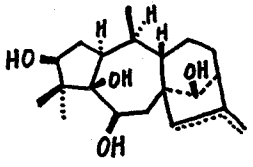
244
[REF] 151
[NC] ring opening with $Pb(OAc)_4$



245
[CN] grayanol B
[REF] 152
[NC] synthesis from **243**

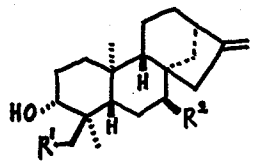


246
[REF] 153
[NC] conversion of **244** to **246** and **247**

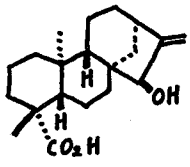


247
[REF] 153
[NC] conversion of **244** to **246** and **247**

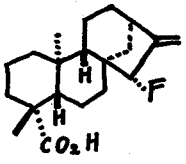
3) Miscellaneous Section



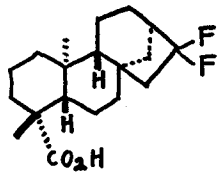
248 $R^1=R^2=H$
249 $R^1=OH$, $R^2=H$
250 $R^1=R^2=OH$
[REF] 154
[NC] microbial transformation of **248–250**



251
[REF] 155
[NC] microbial production of plant gibberellins from **251** in *G. Fujikuroi*

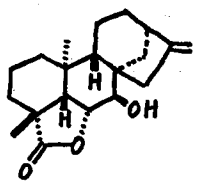


252
[REF] 156
[NC] microbial production of fluoro gibberellins from **252**



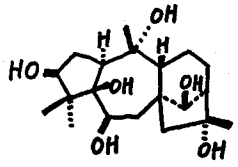
253

[REF] 157
[NC] microbial transformation



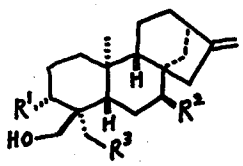
254

[REF] 158
[NC] kaurenolide biosynthesis



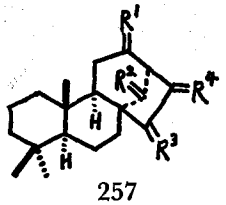
255

[CN] grayanotoxin III
[REF] 159
[NC] biosynthesis of grayanotoxin



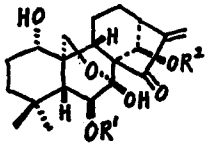
256

[REF] 160
[NC] ^{13}C NMR studies



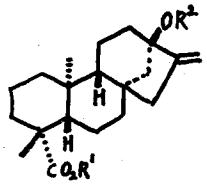
257

[REF] 161
[NC] ^{13}C NMR studies



258

[REF] 162
[NC] antitumor activity of acylated oridonin

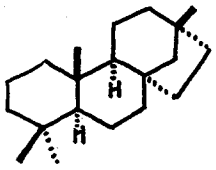


259

[CN] rebandioside-A, D, E
[REF] 163
[NC] structure-sweetness relationship and synthesis

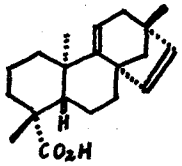
Additional references
[REF] 164
[NC] sensory evaluation of stevioside analogues

X. BEYERANE DERIVATIVES



Beyerane

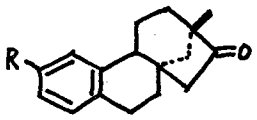
1) Isolation and Structure Determination



260

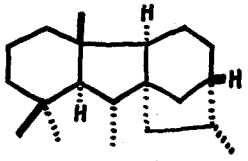
[CN] dehydrostachemic acid
[NS] *Viguiera* species
[REF] 165

2) Synthesis and Reaction



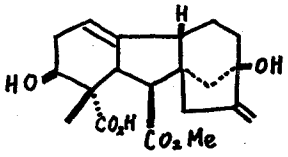
261 R=H
262 R=OMe
[REF] 166

XI. GIBBERELLANE DERIVATIVES

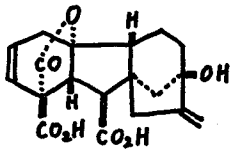


Gibberellane

1) Isolation and Structure Determination



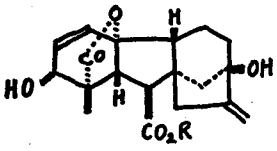
263
[REF] 167
[NC] X-ray analysis



264
[CN] gibberellin A_{59}
[NS] *Canavalia gladiata*
[REF] 168

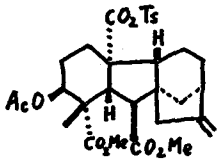
Additional reference
[NS] *Chrysanthemum morifolium*
[REF] 169

2) Synthesis and Reaction

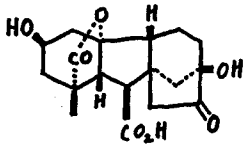


265
[CN] gibberellin A_3 (R=H)
[REF] 170
[NC] degradation

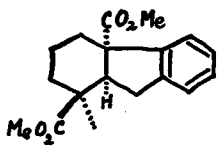
[REF] 171 (R=H)
[NC] chemical conversion to GA_5
[REF] 172 (R=Me)
[NC] reaction with PPh_3
[REF] 173 (R=Me)
[NC] chemical conversion to GA_9 and GA_{20}



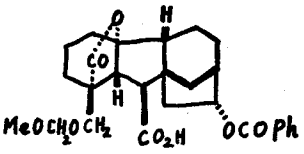
266
[CN] dimethyl ester of gibberellin A_{13} 20-toluene-*p*-sulfonyl anhydride
[REF] 174
[NC] methanolysis, reduction with $NaBH_4$



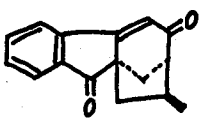
267
[CN] gibberellin A_{29}
[REF] 175
[NC] deuterated and tritiated GA_{29}
[REF] 176
[NC] chemical conversion



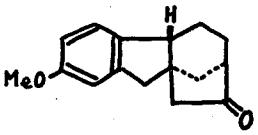
268
[REF] 177
[NC] synthesis



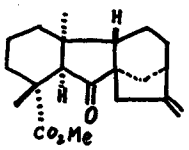
269
[REF] 178
[NC] synthesis



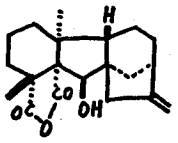
270
[REF] 179
[NC] synthesis



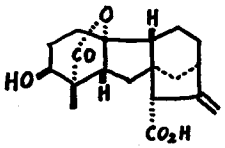
271
[REF] 180
[NC] synthesis



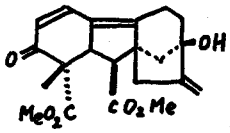
272
[REF] 144
[NC] preparation from fujenal



273
[REF] 144
[NC] preparation from fujenal



274
[CN] 7 (6→15βH)*abeo*-gibberellin A_4
[REF] 181
[NC] preparation form GA_4



275
[REF] 182
[NC] photosensitized dimerization
[REF] 183
[NC] photoreduction
[REF] 184
[NC] photocycloaddition

Additional references

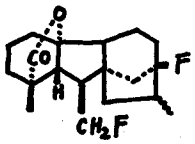
[REF] 185
[NC] chlorination

[REF] 186
[NC] glucosylation

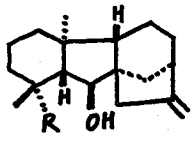
[REF] 187
[NC] methylenation

[REF] 188
[NC] reduction with K-selectride

3) Miscellaneous Section



276
[REF] 189
[NC] feeding experiment



277 R=COOH
278 R=CH_2OH
[REF] 145
[NC] inhibitors of gibberellin biosynthesis

Additional references

[REF] 109
[NC] bioassay

[REF] 157
[NC] biotransformation of fluorogibberellins

[REF] 156
[NC] biotransformation of fluorogibberellins

[REF] 155
[NC] microbial production of plant gibberellins

[REF] 191
[NC] biosynthetic studies

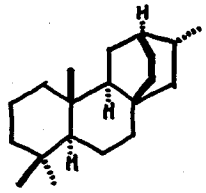
[REF] 192
[NC] biosynthesis in *Phaseolus coccineus*

[REF] 193
[NC] effect on the RNA-ase activity

[REF] 194
[NC] gibberellins in callus of crown gall

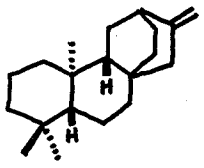
[REF] 195
[NC] chromatographic separation

XII. ATISANE DERIVATIVES

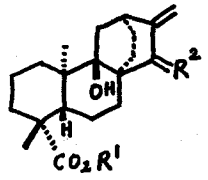


Atisane

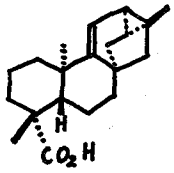
1) Isolation and Structure Determination



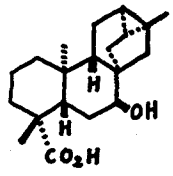
279
[CN] atisirene
[NS] *Thymus capitatus*
[REF] 196



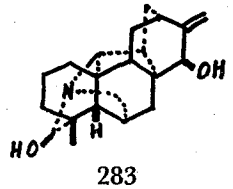
280
[CN] pteroatisene P_1 (R^1=H, R^2=O)
pteroatisene P_2 (R^1=H, R^2=α-H, β-OH)
pteroatisenoside P_1 (R^1=β-D-glucosyl, R^2=O)
[NS] *Pteris purpureorachis*
[REF] 197, 120
[NC] X-ray analysis



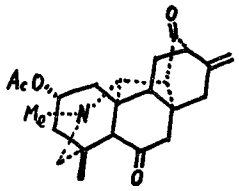
281
[CN] 9,11-dehydro trachylobanic acid
[NS] *Viguiera* species
[REF] 165



282
[CN] ciliaric acid
[NS] *Helianthus grosseserratus*
[REF] 117



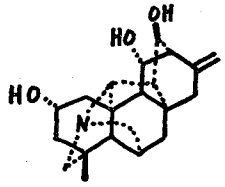
283
[CN] talatisine
[REF] 198
[NC] X-ray analysis



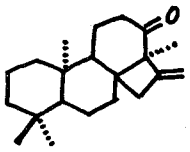
284

[CN] heterophylloidine
[NS] *Aconitum heterophylloides*
[REF] 199
[NC] X-ray analysis on a derivative

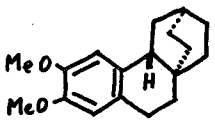
2) Synthesis and Reaction



285
[CN] hetisine
[REF] 200
[NC] rearrangement with acids

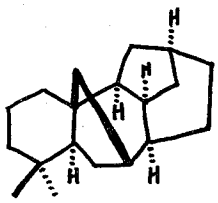


286
[REF] 201
[NC] rearrangement to the atisane skeleton



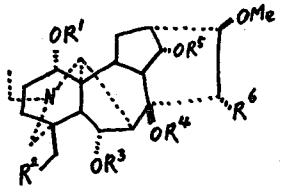
287
[REF] 202
[NC] synthesis from a benzylisoquinoline derivative

XIII. ACONANE DERIVATIVES



Aconane

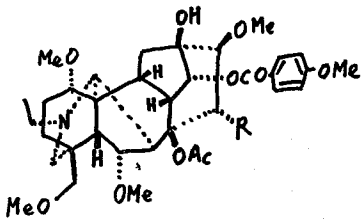
1) Isolation and Structure Determination



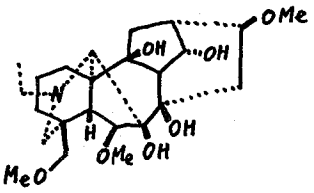
288 $R^1=R^2=R^5=R^6=H$, $R^3=Ac$, $R^4=Me$
[CN] alkaloid A
[NS] *Delphinium bicolour*
[REF] 203
[NC] X-ray analysis

289 $R^1=R^3=Me$, $R^2=OMe$, $R^4=Ac$, $R^5=CO-C_6H_4-OMe$, $R^6=H$
[CN] foresaconitine
[NS] *Aconitum forestii*
[REF] 204

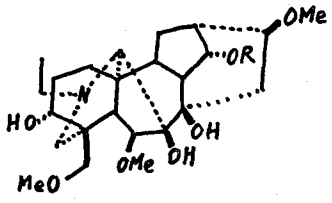
290 $R^1=R^4=R^5=H$, $R^2=OMe$, $R^3=Me$, $R^6=OH$
[CN] 15α-hydroxyneoline
[NS] *Aconitum* species
[REF] 205



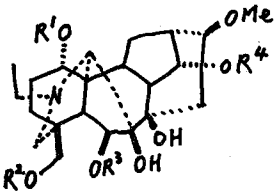
291 R=H
[CN] crassicauline A
[NS] *Aconitum crassicaule*
[REF] 206
292 R=OH
[CN] deoxyjesaconitine
[NS] *Aconitum subcuneatum*
[REF] 207



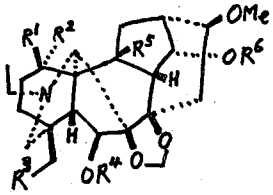
293
[CN] delcaroline
[NS] *Delphinium carolinianum*
[REF] 208

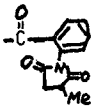


294 R=Me (acomonine)
295 R=H (iliensine)
296 R=-CO- 
(14-benzoyliliensine)
[REF] 209
[NC] structure reinvestigation



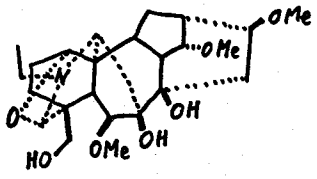
297 $R^1=R^3=R^4=Me$, $R^2=H$ (lycoctonine)
298 $R^1=R^2=R^3=Me$, $R^4=H$ (browniine)
[REF] 210
[NC] revision of the stereochemistry at C(1) for 37 alkaloids
299 $R^1=R^3=R^4=H$, $R^2=Me$
[CN] delphinifoline
[NS] *Aconitum delphinifolium*
[REF] 211
[NC] X-ray analysis
300 $R^1=R^2=R^3=Me$,
$R^4=COCH(CH_3)CH_2CH_3$ (glaucedine)



301 $R^1=R^4=Me$, $R^2=$ 
$R^3=H$(glaudelsine)
[NS] *Delphinium glaucescens*
[REF] 212

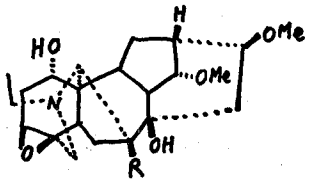
302 $R^1=R^3=OMe$, $R^2=R^4=R^5=R^6=H$ (delcorine)
303 $R^1=R^3=OMe$, $R^2=R^4=R^5=H$, $R^6=Me$ (delcoridine)
[REF] 213

304 $R^1=R^3=H$, $R^2=OMe$, $R^4=Ac$, $R^5=OH$, $R^6=CO\text{-}CH(CH_3)CH_2CH_3$ (glaucenine)
305 $R^1=R^3=H$, $R^2=OMe$, $R^4=Ac$, $R^5=OH$, $R^6=COCHMe_2$ (glaucerine)
306 $R^1=R^3=H$, $R^2=OMe$, $R^4=Ac$, $R^5=OH$, $R^6=COPh$ (glaucephine)
[NS] *Delphinium glaucescens*
[REF] 212



307

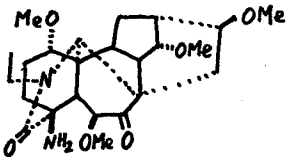
[CN] 18-hydroxy-14,O-methylgadesine
[NS] *Consolida orientalis*
[REF] 214
[NC] X-ray analysis



308 R=H (monticamine)
309 R=OH (monticoline)
[NS] *Aconitum monticola*
[REF] 215

Additisnal reference
[REF] 216
[NC] diterpene alkaloids from
Delphinium cardiopetalum

2) Shythesis and Reaction



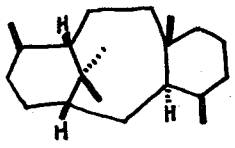
310

[REF] 217
[NC] nitrous acid deamination

3) Miscellaneous Section

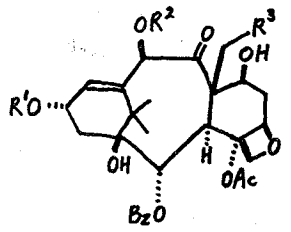
[REF] 218
[NC] ^{13}C NMR spectra

XIV. TAXANE DERIVATIVES



Taxane

1) Isolation and Structure Determination



311 R^1=H, R^2=Ac, R^3=OH
(19-hydroxybaccatin IV)

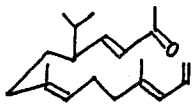
312 R^1=CO—CH(Ph)CHNHCO—C(Me)=CH(Me), CH(OH)
R^2=R^3=H
(10-deacetylcephalomannine)

313 R^1=COCH(OH)CH(Ph)NHBz, R^2=R^3=H
(10-deacetyltaxol)
[NS] *Taxus wallichiana*
[REF] 219
[NC] antitumor diterpenoid

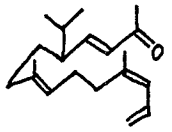
314 R^1=COCH(OH)CH(Ph)NHCOC(Me)=CH(Me),
R^2=Ac, R^3=H
[CN] cephalomannine
[REF] 220
[NC] antileukemic diterpenoid, X-ray analysis

XV. THE OTHERS

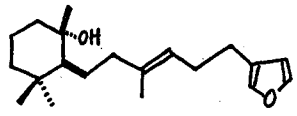
1) Isolation and Structure Determination



315

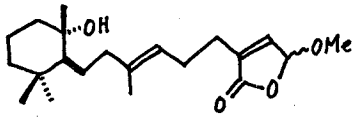


316
[REF] 221
[NC] from cigarette smoke

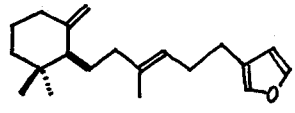


317
[CN] ambliol-A

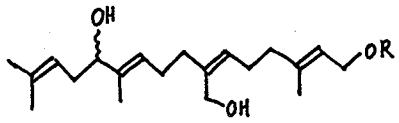
318
[CN] ambliofuran



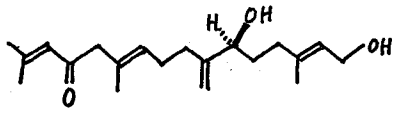
319
[CN] ambliolide



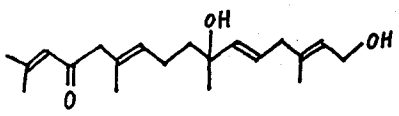
320
[CN] dehydroambliol-A
[NS] *Dysidea ambia*
[REF] 45
[NC] marine sponge metabolites



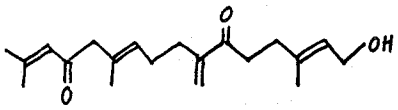
321 R=H
322 R=Ac
[NC] *Grazielia* species
[REF] 15



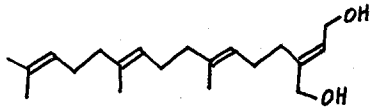
323



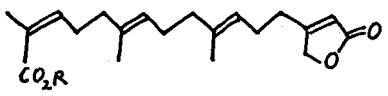
324



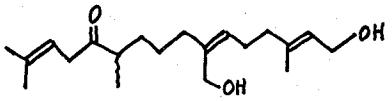
325
[NS] *Cystoseira crinita*
[REF] 222
[NC] from brown alga



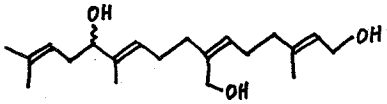
326
[NS] *Kingianthus paradoxus*
[REF] 223



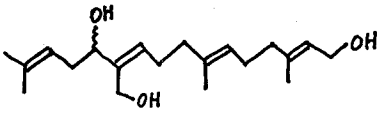
327 R=H
[CN] dimeroperatic acid
328 R=Me
[CN] methyl dimeroperatate
[NS] *Dimerostemma asperatum*
[REF] 224



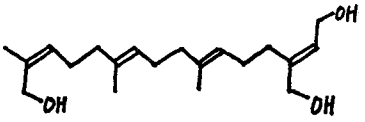
329



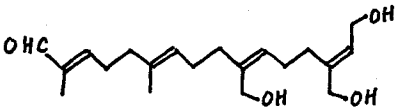
330
[NS] *Lasiolaena santosii*
[REF] 20



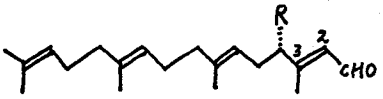
331
[NS] *Zinnia tenuiflora*
[REF] 225



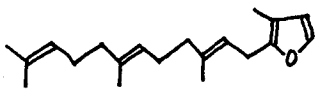
332
[NS] *Bejaranoa semistriata*
[REF] 226



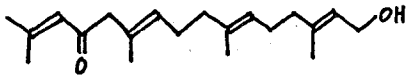
333
[NS] *Mikania officinalis*



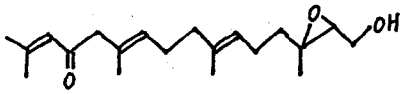
334 R=H
335 R=H (2,3 Z)
336 R=OH



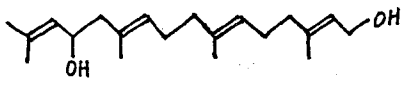
337
[NS] *Mikania sessilifolia*
[REF] 227



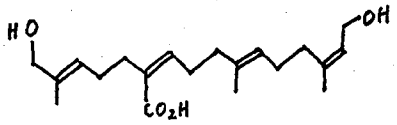
338
[CN] eleganolone



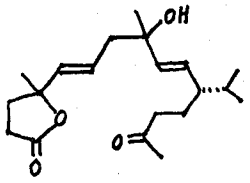
339
[CN] epoxyeleganolone



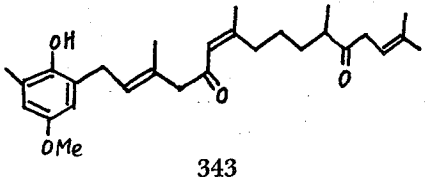
340
[CN] elegandiol
[NS] *Cystoseira elegans*
[REF] 228
[NC] seasonal variation of a cyclic diterpenes



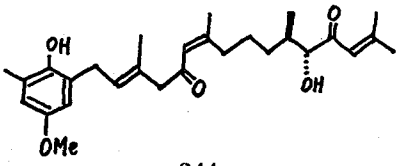
341
[NS] *Wyethia helenioides*
[REF] 229



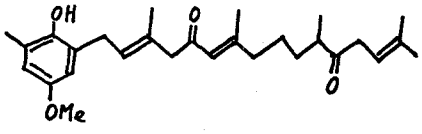
342
[REF] 230
[NC] from burley tobacco



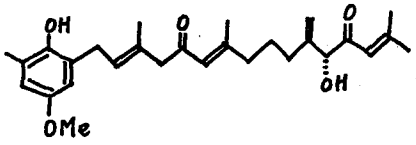
343
[CN] 5′, 12′-dioxohalidrol



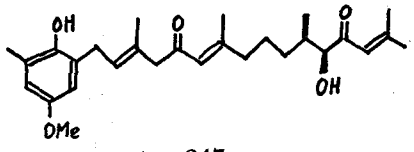
344
[CN] 12′α-hydroxy-5′, 13′-dioxohalidrol



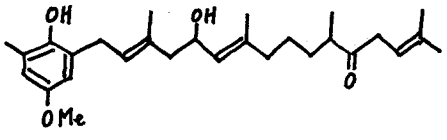
345
[CN] 5′, 12′-dioxoisohalidrol



346
[CN] 12′α-hydroxy-5′, 13′-dioxoisohalidrol

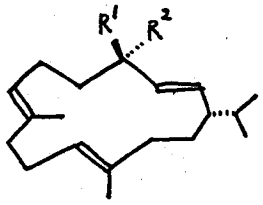


347
[CN] 12′β-hydroxy-5′, 13′-dioxoisohalidrol

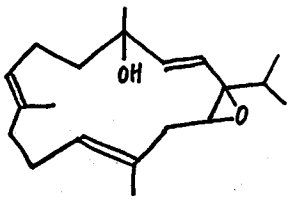


348
[CN] 5′-hydroxy-12′-oxohalidrol

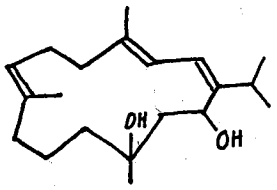
[NS] *Halidrys siliquosa*
[REF] 231



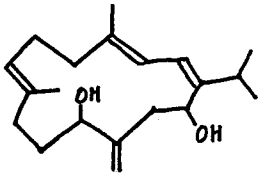
349 R^1=OH, R^2=Me
[CN] thunbegol (isocembrol)
350 R^1=Me, R^2=OH
[CN] 4-epiisocembrol
[NS] *Pseudotsuga menziesii*
[REF] 232



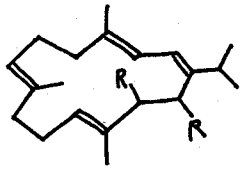
351
[CN] sarcophytol-C
[NS] *Sarcophyton glaucum*
[REF] 233



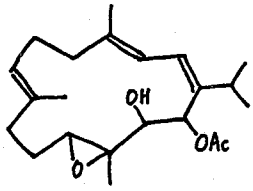
352
[CN] sarcophytol-D



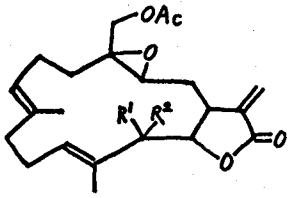
353
[CN] sarcoahytol-E
[NS] *Sarcophyton glaucum*
[REF] 233



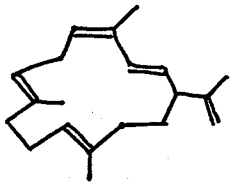
354 R=H
[CN] cembrene-C
355 R=OH
[CN] sarcophytol-B
[NS] *Alcyonium flaccidum*
[REF] 234



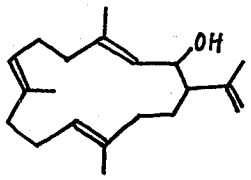
356
[CN] flaccidoxide
[NS] *A. flaccidum*
[REF] 234



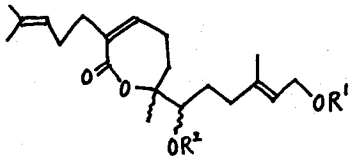
357 R^1=R^2=H
358 R^1=H, R^2=OH
359 R^1=OH, R^2=H
[NS] *Lobophytum crassum*
[REF] 234



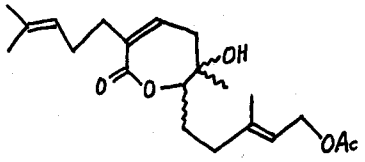
360
[CN] cembrenene



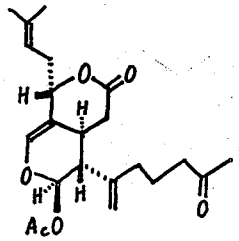
361
[CN] mayol
[NS] *Sinuralia mayi*
[REF] 235



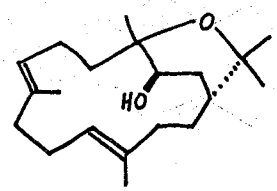
362 R^1=R^2=H
[CN] acanthoaustralide
363 R^1=Ac, R^2=H
[CN] acanthoaustralide-1-O-acetate
[NS] *Acanthospermum australi*
[REF] 236



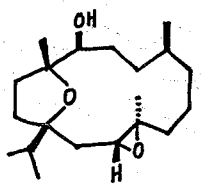
364
[CN] isoacanthoaustralide-1-O-acetate
[NS] *A. australe*
[REF] 236



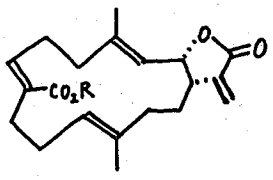
365
[CN] alcyonolide
[NS] *Akyonimu* species
[REF] 237



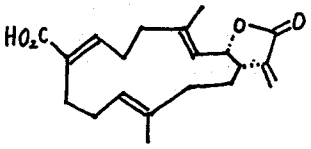
366
[CN] decaryiol
[NS] *Sarcophyton decaryi*
[REF] 238



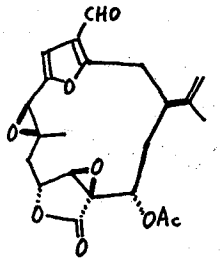
367
[CN] incensole oxide
[REF] 239
[NC] X-ray analysis



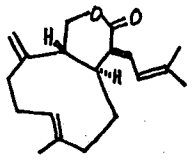
368
[CN] lobohedleolide
[NS] *Lobophytum hedleyi*
[REF] 240
[NC] X-ray analysis; growth inhibition of the Hella cells



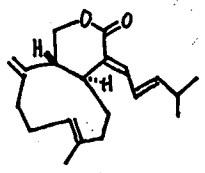
369
[CN] (7Z)-lobohedleolide
[NS] *L. hedleyi*
[REF] 240



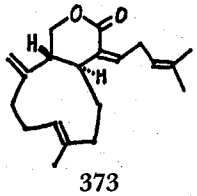
370
[CN] lophotoxin
[NS] *Lophogorgia alba, L. cuspidata, L. rigida, L. chilensis*
[REF] 241
[NC] new neuromuscular toxin



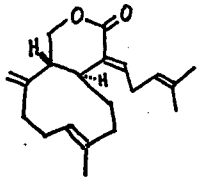
371
[CN] coraxeniolide-A



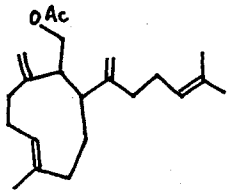
372
[CN] coraxeniolide-B



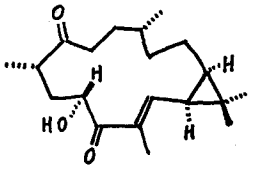
373
[CN] coraxeniolide-C



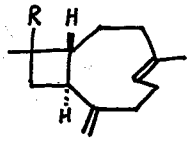
374
[CN] coraxeniolide-C′



375
[CN] corabohcin
[NS] *Corallium* species
[REF] 242



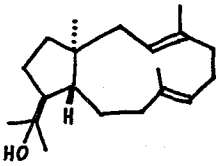
376
[CN] crotonitenone
[NS] *Croton nitens*
[REF] 243



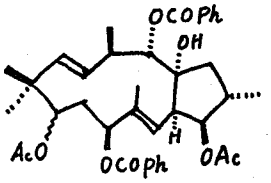
377 R=$CH_2CH_2CH(O)CMe_2$

378 R=$CH_2CH_2CH(OH)CMe_2Cl$

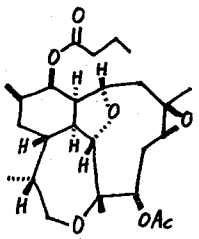
[NS] *Nephthea chabrolli*
[REF] 244



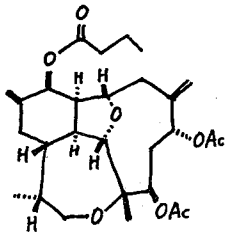
379
[CN] 18-hydroxy-3,7-dolablladiene
[NS] *Dictyota dichotoma*
[REF] 245



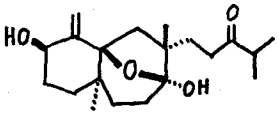
380
[NC] euphornin
[NS] *Euphorbia maddeni*
[REF] 246



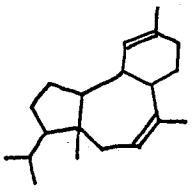
381



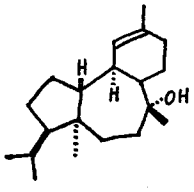
382
[NS] *Briareum asbestinum*
[REF] 247
[NC] from toxic extracts



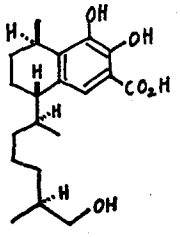
383
[CN] linearol
[NS] *Dictyota linearis*
[REF] 248



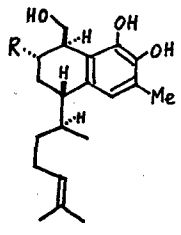
384
[CN] sphaerodiene



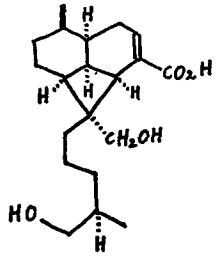
385
[CN] presphaerol
[NS] *Sphaerococcus coronopifolius*
[REF] 249
[NC] structure **385** (reassigned by X-ray analysis)



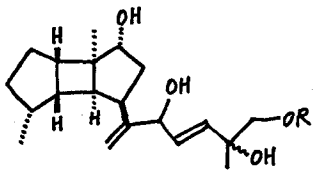
386



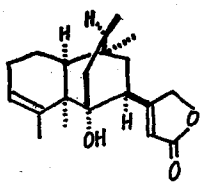
387 R=H
388 R=OH
[NS] *Eremophila* species
[REF] 250, 251
[NC] X-ray analysis



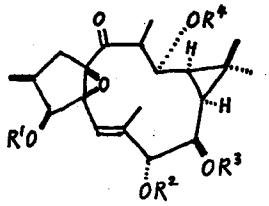
389
[NS] *Eremophila decipiens*
[REF] 252
[NC] chemical degradation



390 R=H, **391** R=Ac
[NS] *Stoechospermum marginatum*
[REF] 253

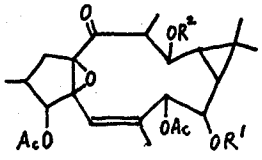


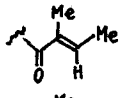
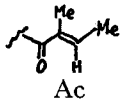
392
[CN] tricyclosolidagolactone
[NS] *Solidago altissima*
[REF] 254
[NC] X-ray analysis



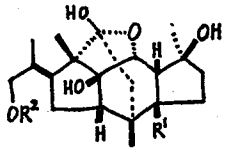
	R^1	R^2	R^3	R^4
393	Ac	Ac	benzoate	Ac
394	Ac	Ac	angelate	Ac
395	Ac	Ac	tigliate	Ac
396	H	Ac	benzoate	Ac
397	H	Ac	angelate	Ac
398	H	H	tigliate	Ac
399	H	H	H	Ac
400	H	H	H	H
401	**Ac**	**Ac**	**Ac**	**Ac**

[NS] *Euphorbia kamerunica*
[REF] 255

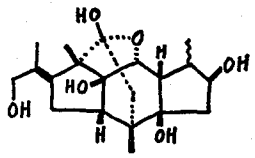


	R^1	R^2
402	Ac	
403	H	
404	Ac	Ac

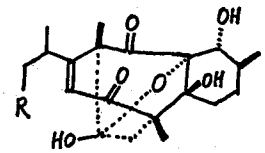
[NS] *Euphorbia kamerunica*
[REF] 256
[NC] from the cytotoxic fraction



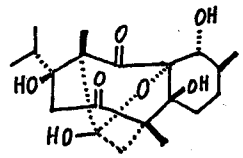
405 $R^1=R^2=H$
[CN] cinncassiol D_1
406 $R^1=H$, $R^2=\beta$-D-glc·pyr-
[CN] cinncassiol D_1 glucoside
407 $R^1=OH$, $R^2=H$
[CN] cinncassiol D_2
408 $R^1=OH$, $R^2=\beta$-D-glc·pyr-
[CN] cinncassiol D_2 glucoside



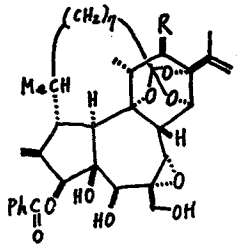
409
[CN] cinncassiol D_3
[NS] *Cinnamomi cortex*
[REF] 257
[NC] from the fraction exhibiting anti-complement activity; X-ray analyses



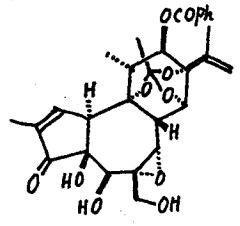
410 R=O-β-D-glc·pyr
[CN] cinncassiol C_1 glucoside
411 R=H
[CN] cinncassiol C_2



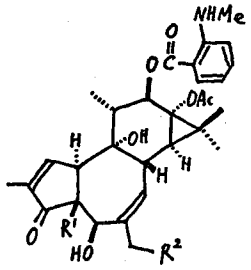
412
[CN] cinncassiol C_3
[NS] *Cinnamomi cortex*
[REF] 258
[NC] from the fraction exhibiting anticomplement activity



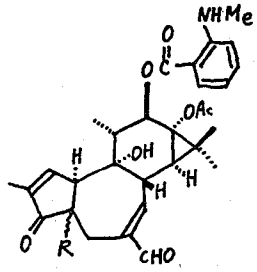
413 R=OAc
[CN] linifolin A
414 R=H
[CN] linifolin B
[NS] *Pimelea linifolia*
[REF] 259
[NC] fish toxin



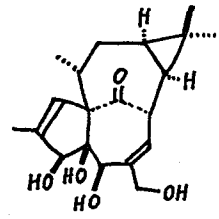
415
[CN] genkwadaphnin
[NS] *Daphne genkwa*
[REF] 260
[NC] new antileukemic principle



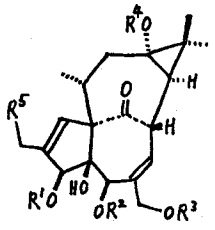
416 R^1=H, R^2=OH
[CN] sapintoxin B
417 R^1=R^2=H
[CN] sapintoxin C
418 R^1=R^2=OH
[CN] sapintoxin D
[NS] *Sapium indicum*
[REF] 261, 262



419 R=β-H
420 R=α-H
[NS] *Sapium indicum*
[REF] 263
[NC] first natural tiglianes to exhibit a C-20 aldehyde



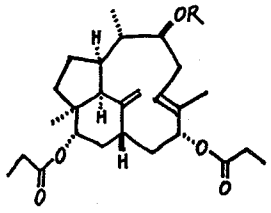
421
[CN] ingenol
[NS] *Euphorbia ingens*
[REF] 264



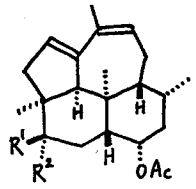
422 $R^2=R^3=R^5=H$, $R^1=A$. $R^4=B$
423 $R^1=R^3=R^5=H$, $R^2=A$, $R^4=B$
424 $R^1=R^2=R^5=H$, $R^3=A$, $R^4=B$
425 $R^1=A$, $R^2=R^3=R^5=H$, $R^4=C$
426 $R^1=R^4=A$, $R^2=R^3=H$, $R^5=D$
427 $R^1=R^3=H$, $R^2=R^4=A$, $R^5=D$
428 $R^1=R^2=H$, $R^3=R^4=A$, $R^5=D$

[NS] *Euphorbia cyparissias*
[REF] 265
[NC] highly irritant activity

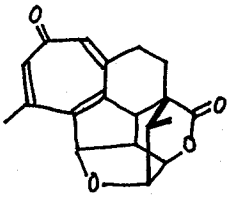
A=CO-CH(Me)-CH(Me)-Me
B=$COC_{11}H_{23}$
C=COC_9H_{19}
D=OCO-CH(Me)CH(Me)-Me



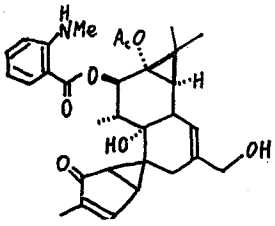
429 R=COEt
430 R=COMe
[NS] *Nasutitermes* species
[REF] 266
[NC] termite soldier defense secretion



431 $R^1=R^2=O$
432 $R^1=OAc$, $R^2=H$
433 $R^1=H$, $R^2=OAc$
[NS] *Bulbitermes singaporensis*
[REF] 267
[NC] defense secretion of the nasute termite



434
[CN] harringtonolide
[NS] *Cephalotaxus harringtonia*
[REF] 84
[NC] podolactone-type plant growth inhibition



435
[CN] sapintoxin A
[NS] *Sapium indicum*
[REF] 268
[NC] irritant compound

Additional references

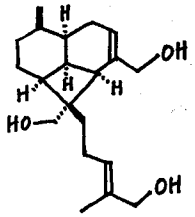
[REF] 12
[NC] cembrane diterpenoids from oleoresin of *Picea-ajanensia*

[REF] 269, 270
[NC] phytochemical cultivation of Brazilian velloziaceae

[REF] 271
[NC] diterpenoids of oleoresin of far east *Abies* species

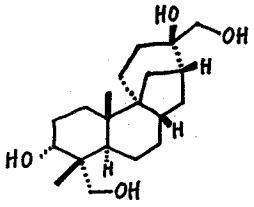
[REF] 272
[NC] new diterpenic acetate from a *Sideritis pusilla*

2) Synthesis and Reaction



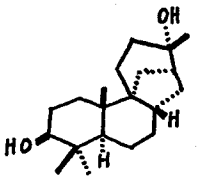
436

[CN] (+)trihydroxydeci-piadiene
[REF] 273
[NC] total synthesis



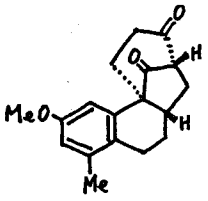
437

[CN] (+)aphidicolin
[REF] 274, 275
[NC] total synthesis



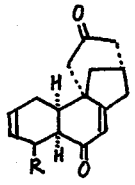
438

[CN] (+)maritimol
[REF] 276
[NC] total synthesis

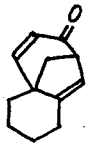


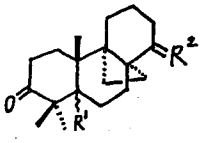
439

[REF] 277
[NC] synthesis of the basic skeleton of aphidicolan-type diterpenes

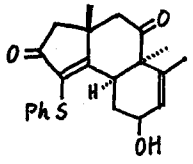


440 R=H, 441 R=OAc
[REF] 278
[NC] synthesis of the ring skeleton of aphidicolin and the related natural products



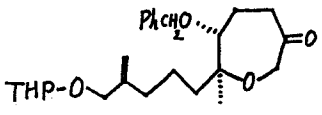


442 443 $R^1=\alpha$-H, R^2=O
444 $R^1=\beta$-H, $R^2=\alpha$-OH, β-H
[REF] 279
[NC] preliminary studies on the synthesis of aphidicolin



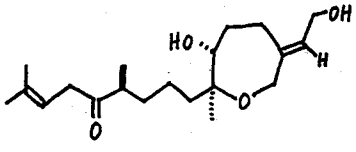
445

[REF] 280
[NC] synthesis of a potential intermediate directed towards cyathins



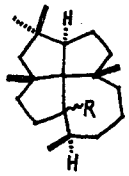
446

[REF] 281
[NC] stereoselective synthesis of a key intermediate for the total synthesis of (+)zoapatanol



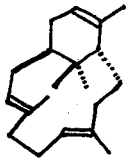
447

[CN] (+)zoapatanol
[REF] 282
[NC] toatl synthesis

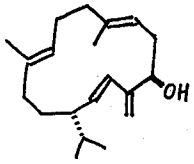


448 R=α-H
[CN] laurenane

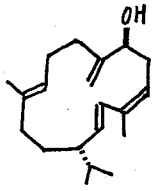
449 R=β-H
[CN] 1βH-laurenane
[REF] 283



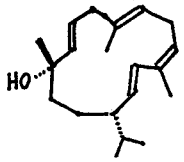
450
[REF] 284
[NC] synthesis of a geometrical isomer of anhydroverticillol



451



452



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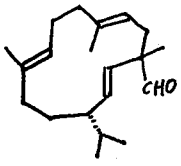
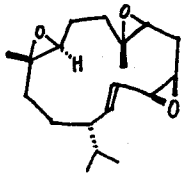
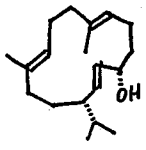


454

[REF] 285
[NC] photooxidation products of cembrene

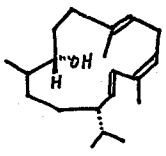


455
[REF] 286
[NC] complete epoxidation product of cembrene; X-ray analysis

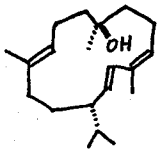


456
[CN] cembranol

453

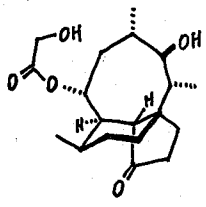


457



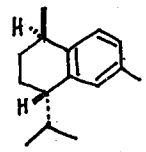
458

[REF] 287
[NC] reduction products of 11*S*,12*S*-epoxycembrene or 7*S*,8*S*-epoxycembrene by $LiAlH_4$; growth inhibition of red wheat



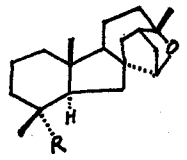
459

[REF] 288
[NC] chemical conversion from a pleuromutilin derivative

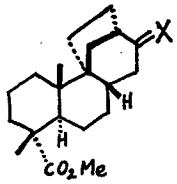


460

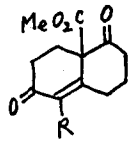
[CN] (1*R*,4*S*)-(−)-calamenene
[REF] 289
[NC] synthesis from dihydroxyserrulatic acid



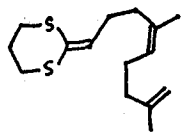
461 R=Me
462 R=CH_2OCOPh
463 R=CO_2Me
[REF] 290
[NC] chemical conversion (*in vitro* biosynthetic studies); X-ray analysis



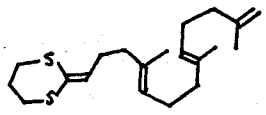
464 X=α-Me, β-OMe
465 X=α-OMe, β-Me
[REF] 291
[NC] cyclization products of methyl isopimarate and pimarate



466 R=H
467 R=Me
[CN] Wieland-Mischer ketone analogues
[REF] 292
[NC] intermediates in the synthesis of bruceantin



468



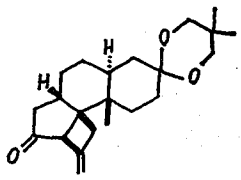
469

[REF] 293
[NC] synthesis of *Z*- and *E*-polyunsaturated isoprenoids

Additional reference

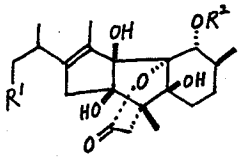
[REF] 294
[NC] regiospecific and stereoselective 3α-hydroxylation of fusicoccin derivatives

3) Miscellaneous Section

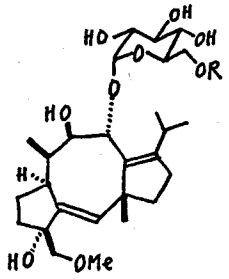


470

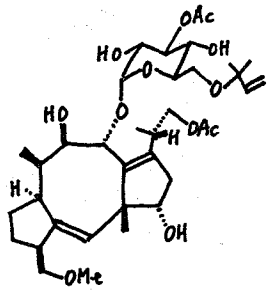
[REF] 295
[NC] X-ray analysis of a stemodin intermediate **470**



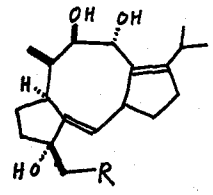
471 R^1=H, R^2=Ac
472 R^1=R^2=H
473 R^1=OH, R^2=H
474 R^1=OAc, R^2=H
475 R^1=O-glc(Ac), R^2=H
[REF] 296
[NC] EI mass spectra of *Cassia* diterpenes



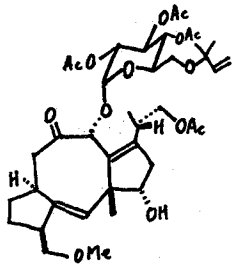
476 R=Me
[CN] cotylenin-E
477 R=H
[CN] cotylenin-J
[REF] 297
[NC] germination-stimulating activity



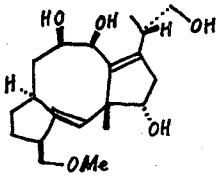
478
[CN] fusicoccin-A
[REF] 297
[NC] germination-stimulating activity



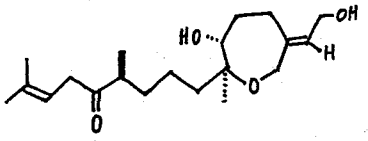
479 R=OMe
[CN] cotylenol
480 R=OH
[CN] 16-O-demethyl-cotylenol
[REF] 297
[NC] germination-stimulating activity



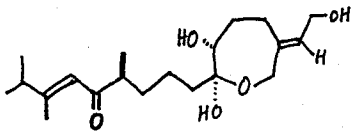
481
[CN] fusicoccin triacetate
[REF] 298
[NC] structure revision (**481**); preparation of **482**



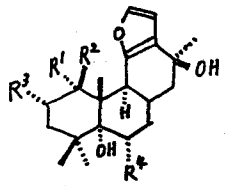
482
[CN] 9-epifusicoccin aglycone
[REF] 298
[NC] structure revision (**481**); preparation of **482**



446
[CN] zoapatanol
[REF] 299
[NC] ^{13}C NMR studies



483
[CN] montanol
[REF] 299
[NC] ^{13}C NMR studies



484 $R^1=R^3=OAc$, $R^2=R^4=H$
485 R^1, $R^2=O$, $R^3=H$, $R^4=OAc$
486 R^1, $R^2=O$, $R^3=H$, $R^4=OH$
487 R^1, $R^4=OH$, $R^2=R^3=H$
[RER] 300
[NC] ^{13}C NMR studies of cassane diterpenoids; stereochemistry of the caesalpins

Additional reference

[REF] 301
[NC] computer-assisted structural interpretation of ^{13}C NMR spectral data

XVI. REVIEW ARTICLES

[REF] 302
[NC] irritant and defense substances

[REF] 303
[NC] terpenoid metabolites of mushrooms

[REF] 304
[NC] naturally occuring gibberellins (in Japanese)

[REF] 305
[NC] biologically active glucosides produced by fungi (in Japanese)

[REF] 306
[NC] fungal elicitors of the phytoalexin response

[REF] 307
[NC] chemistry of new natural sweet-principles (in Japanese)

[REF] 308
[NC] cocarcinogenesis and tumor promoters

[REF] 309
[NC] Chinese medicinal plants (in Japanese)

[REF] 310
[NC] decomposition of α-diazo ketones

[REF] 311
[NC] singlet oxygen in organic synthesis

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